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Biodegradability and Tensile Properties of Compatibilised Polyethylene/Rice Bran Film

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In this study, a degradable film based on polyethylene (PE) and Rice bran (RB) as filler were developed. Low density polyethylene (LDPE) and RB (0 - 5 %) were prepared by using twin screw extruder and blown film machine. The effect of 1 parts per hundred (phr) of Ultra PlastTM10 (TP10) compatibiliser on the biodegradability, tensile properties and morphological properties of LDPE/RB films was evaluated. The presence of high RB contents had an adverse effect on the tensile strength of LDPE/RB films. Incorporation of compatibiliser increased the tensile strength of LDPE/RB films. The addition of TP10 in the films has enhanced the interfacial adhesion, distribution and dispersion phase and thus, improved the tensile properties of the films. The addition of RB in LDPE/RB films has increased their biodegradability due to the attacks of microorganism in the soil. Incorporation of TP10 in LDPE/RB films led to a negative effect on the rate of biodegradation.

1. Introduction

Plastics waste is a significant portion of the total municipal solid waste. The difficulty to manage the plastic waste arise due to a longer time to degrade or cannot degrade properly. Plastic waste can also degrade into substances that are hazardous and polluting. It adversely affects the environmental, economic and ecological system, especially to a human being. The recycling and incineration methods had been done to minimise these problems. Only a small percentage of plastic waste is recyclable and most of them end up in the municipal landfill. The emission of corrosive gas, high capital cost, toxic gases and operating at high temperature make incineration less attractive (Moghadam et al., 2013). One of the solutions to tackle the problem of plastic waste management is the production and use of environmental friendly degradable polymers, especially in the packaging applications (Monica et al., 2015). The environmentally degradable polyelfin films are defined as those materials that contain degradation process of polyolefin article (bag/film/sheet) under conditions of composting. There are many renewable sources that are used to produce biodegradable polymer that is comparable to substitute the growing use of non-biodegradable polymer (Tudorachi et al., 2000).

Most countries use rice as staple food especially Asian countries. The major rice growing countries are India, Thailand, Indonesia, China, Bangladesh, Vietnam, Burma, Philippines and the Japan (Hu et al., 2009). RB is a by-product of the rice milling process during the spreading of brown rice to white rice. The outer brown layer was called RB and is composed of the rice germ and several sub-layers, which account for approximately 8 % by weight of paddy rice and contain over 60 % of the nutrients found in each kernel of rice.

Today, the challenge of producing biodegradable polymer becomes the focus of research interest in order to overcome the problem of plastic waste management (Seggiani et al., 2015). Many researches have been carried out to produce the degradable plastic film from LDPE and product from natural resources such as starch (Garg and Jana, 2006), flour (Morreale et al., 2008) and chitosan (Bourtoom and Chinnan, 2008). In this study, the RB from local producer will be used to produce degradable LDPE/RB film. The effect of RB content and the addition of compatibiliser on the tensile properties, biodegradability and morphology will be evaluated. From the literature search, there is no report on the effect of TP10 as compatibiliser on the

919

properties of LDPE/RB film. The outcome of this study will be able to enhance the properties of this product and helps the producer to overcome few of their challenges in producing biodegradable film.

2. Experimental

2.1 Materials

LDPE (LDF260GG; density: 0.922 g/cm³) was purchased from TITAN Chemical, Malaysia. RB with a particle size range of 100 - 500 μ m was purchased from local rice mills in Kelantan, Malaysia. Ultra PlastTM TP10 (TP10) which were used as a compatibiliser of LDPE/RB films were purchased from Sigma Aldrich Malaysia.

2.2 Experimental Procedure

RB was dried in an oven at 60 °C for 24 h prior to mixing process. LDPE, RB and compatibiliser were melt mixing with different concentration of RB (0 - 5 %) by using twin screw extruder with a temperature range of 150 - 190 °C while the speed was 50 rpm. The compound was pelletized by using pelletiser. The pellet of the compound was blown into a film by using blown film machine (Tai King 42440) with temperature range 115 - 160 °C at the speed of 650 rpm. The tensile properties of a degradable film of LDPE/RB such as tensile strength and percentage of elongation at break were measured by using Lloyd Instruments testing machine. The machine speed was set at 50 mm/min and the sample film was prepared in a rectangular shape as ASTM D882. The morphology of LDPE/RB film was analysed by using scanning electron microscopy (SEM). The soil burial test has been carried out to investigate the biodegradability and the rate of biodegradation of the films that buried in the soil according to ASTM D1435. The samples of the films which was cut into uniform strip sheet (about 50 mm × 10 mm) were buried in the soil. The poly bag containing soil and samples were exposed to environmental effects such as sunlight and rains for several months. The samples were collected following the schedule to determine the rate of biodegradation. The surfaces of the samples were cleaned with a tissue and dried. The weights of all samples were weighed, measured and recorded. The percentage of degradation below;

$$\% \text{ Degradation } = \frac{m_o - m_d}{m_o} \times 100 \%$$
⁽¹⁾

Where m_0 = initial weight of sample and m_d = final weight after buried in soil.

3. Results and Discussion

The effect of TP10 addition on the tensile strength and percentage of elongation at break of LDPE/RB films are presented in Figure 1 and 2. Figure 1 shows the tensile strength of LDPE/RB films with and without TP10 at different RB contents. The tensile strength of LDPE/RB film decreased linearly as the rice bran content was increased. The result shows that the tensile strength of pure LDPE films was 9.43MPa which is higher than other films. With the addition of 2% and 4 % of rice bran, the tensile strength dropped to 6.14MPa and 4.96 MPa, respectively. The tensile strength of LDPE/rice bran films dropped about 34.9% and 47.4%. This is probably due to the increase in brittleness of the films caused by the blending and replacement of the tough matrix (LDPE) with brittle material which is rice bran.





Figure 1: Effect of TP10 on tensile strength for LDPE/RB films and at different RB loadings

Figure 2: Effect of TP10 on elongation at break of LDPE/RB films at different RB loadings

The above phenomenon can be explained by examining their morphology through scanning electron microscope (SEM) as shown in Figure 3 (a-c). The investigation of morphology structure for polymer blends and composite such as films is very important because it enables the determination of the properties of that material such as mechanical properties, adhesive properties and viscoelastic properties (Wang et al., 2005). Figure 3a and 3b show that rice bran was present as filler or non-reinforcing filler because the granular structure of rice bran was neither melted nor destroyed during processing. In this case, the rice bran used in this study contained 20% of rice starch. The distribution and dispersion of 2 wt% rice bran content in LDPE matrix seems to be uniform and homogeneous, however, at 4 wt% concentration, the agglomeration phenomenon occurred and the distribution and dispersion of rice bran in LDPE matrix was found to be not homogeneous. This is probably due to the hygroscopic nature of rice bran. In addition, the agglomeration of rice bran can affect the mechanical properties. Therefore, increasing rice bran content in LDPE films had decreased the homogeneity. The tensile strength of LDPE/RB/TP10 films decreased as the RB content was increased. This is due to the increase in brittleness of the films caused by the replacement of the tough matrix (LDPE) with brittle material which is rice bran. The tensile strength of 1 wt% RB in LDPE/RB/TP10 films was quite similar to the pure LDPE films and the difference is only 0.86 %. Figure 1 shows that, the tensile strength of LDPE/RB/TP10 films is higher than the films without TP10. The incorporation of TP10 in LDPE/RB films has increased the tensile strength of the film. The difference in polarity between LDPE and RB caused incompatible blending between the two phases and produced brittle films. Therefore, TP10 is needed to enhance compatibility and thereby improving the tensile properties.



(a)



Figure 3: SEM micrographs of LDPE/RB films with different ratio of RB in LDPE content: (a) 2 %, (b) 4 % and (c) LDPE

(b)

The compatibility was enhanced by improving the interfacial adhesion between LDPE and RB, reduced the agglomeration phenomenon and improved dispersion of RB particles in LDPE matrix as can be seen in Figure 4(a) to 4(b). The TP10 acted as a compatibiliser in the blended films of RB and LDPE and it also enables preservation of the physical properties of LDPE/RB films. The low molecular weight of TP10 caused viscosity of blending LDPE and RB to reduce during the extrusion processing and thus enhancing their flow and easiness of the process. SEM micrographs of LDPE/RB/TP10 films improved the distribution and dispersion of RB in LDPE matrix compared to LDPE/RB films (Figure 3(a) and 3(b)). The improvement is due to a good compatibility between the RB and LPDE. The particles size is smaller and the dispersion of RB particles in LDPE/RB/TP10 films was more homogeneous compared with the films without TP10. As can be seen in Figure 4(b) the distribution of RB particles for 4 wt% of RB in LDPE/RB/TP10 was better than the LDPE/RB films (Figure 3(b)). This probably causes the increase in the interfacial adhesion between the RB and LDPE matrix. Increased the tensile strength of compatibilised LDPE/RB film.



Figure 4: SEM micrographs of LDPE/RB films with addition TP10: (a) 2 wt% RB, (b) 4 wt% RB Soil burial Test

Figure 2 shows the effect of TP10 on the percentage of elongation at break of LDPE/RB films. The percentage of elongation at break for both formulations reduced as the RB content was increased. The elongation at break of LDPE/RB film decreased as the rice bran content was increased. This is due to the low interfacial adhesion between the two phases. The elongation at break for the pure LDPE film was 136.57 % while at 4 wt% of rice bran the content was 21 %. Therefore, at higher rice bran loading, the elongation at break dropped drastically until 84.62 % compared with the pure LDPE films. Starch granules are highly hydrophilic and contain hydroxyl group while LDPE is a non-polar, therefore incorporation of starch in LDPE content that would cause the stronger formation of interfacial adhesion between starch and LDPE is not feasible. As a result, the percentage of elongation at break decreased as the starch content increased. The filler-filler or particle-particle interaction increased as the rice bran content was increased. It was more obvious on the surface of the film with 4 wt% of rice bran content. The films became more rigid and brittle with increasing rice bran content, thus decreasing the elongation at break. Furthermore, incorporation of starch in LDPE causes the discontinuity in a matrix which gives effect to elongation at break. This is due to the lack of chemical interaction between the two phases. From these results, it can be concluded that the compatibilizer is needed to produce a good distribution and dispersion of rice bran in LDPE matrix to improve the interfacial adhesion as also suggested by another researcher such as Sabetzadeh et al. (2012). TP10 was added into LDPE/RB film in order to improve the compatibility of LDPE and rice bran. The percentage of elongation at break of LDPE/RB/TP10 films was higher than the LDPE/RB films. The result of percentage of elongation at break of LDPE/RB/TP10 films (1 wt% RB) was quite high compared to the pure LDPE films and the difference is 35.3 %. Figure 2 shows that after the addition of TP10 to LDPE/RB film, the percentage of elongation at break increased to 154.7 % for 2 wt% RB and 155.3 % for 4 wt% RB. This is due to the presence of TP10 that improved their flexibility and elasticity of LDPE/RB/TP10 films. The incorporation of TP10 in LDEP/RB films. has changed and improved their homogeneous phase. It improved the distribution and dispersion of RB

922

particles in the LDPE matrix as shown in Figure 3. According to Rahman et al. (2010), high energy or load was required to break or fracture the rice husk/HDPE/TP10 blends during deformation process.

Soil burial test was carried out to determine the effect of RB content on the biodegradability properties of LDPE/RB films based on the percentage weight change or loss. All the samples of LDPE/RB films were buried in the soil and left exposed to the environment (open environment). The effect of compatibiliser on the biodegradability of LDPE/RB films had been analysed and presented in this section. Figure 5 shows the effect of time of burial to the percentages of weight loss for 2 wt% and 4 wt% of LDPE/RB films and LDPE/RB films with TP10. The RB content had influenced the percentages of weight loss of LDPE/RB films. The percentage of weight loss increased with increasing RB content in LDPE/RB films. Similar finding was reported by Soni and Saiyad (2011). In this case, the microorganism of soil would attack the polymer or LDPE strips. These microorganisms possibly attack the RB content in LDPE matrix and this has caused the LPDE chains to fracture or break. According to Borghei et al. (2010), the incorporation of starch in LDPE matrix makes the microorganism in the soil to attack their blends. The capability of RB (hydrophilic) in LDPE films to absorb water influenced their physicochemical properties and the tendency to biodegradation and hydrolysis increased. As well known, the plastic such as LPDE is resistant to degradation in natural environment because the microorganisms do not have the enzymes to degrade the LDPE or synthetic polymer. The hydrophobic LDPE caused the enzymes activities to become limited. The presence of hydrophilic RB in LDPE films indirectly improved their ability of degradation and biodegradability. Similar discovery was also reported by Borghei et al. (2010) and Khoramneiadian (2011).

As observed in Figure 5, incorporation of TP10 as compatibiliser in LDPE/RB films slightly decreased their rate of biodegradation compared with LDPE/RB films. As stated in earlier discussion, the incorporation of compatibiliser in LDPE/RB films improved the interfacial adhesion between two phases which effect on the rate of biodegradation.



Figure 5: Effect of time of burial on the percentages of weight loss of LDPE/RB films

4. Conclusion

It can be concluded that RBRB content strongly affected the tensile properties, morphological and biodegradability properties of LDPE/RB film. As a conclusion, the addition of RB content in LDPE films has decreased the tensile properties such as tensile strength and elongation at break of the films. The maximum composition of RB loading can be incorporated to produce LDPE films by about 5 % with and 4 % without the addition of compatibiliser. The addition of 1 parts per hundred (phr) TP10 in the LDPE/RB films has increased the physical properties of LDPE/RB films. In addition, the low molecular weight of TP10 caused the viscosity of blended LDPE and RB to reduce during the extrusion process and thus enhancing the flow and easiness of the process. SEM images revealed that the Incorporation of TP10 also improved the interfacial adhesion between LDPE and RB, thus reducing the agglomeration phenomenon and improved dispersion of RB particles in LDPE matrix thus improve the tensile properties of LDPE/RB film. The addition of RB in LDPE/RB films has increased the biodegradability of the film due to the hydrophilic nature of RB which increased the rate of degradation. The present study provides the valuable information on the underlying behaviour of the RB in the fabricated degradable LDPE/RB film. These findings enable the researchers to know more about the

nature of RB, which, in turns, facilitates the future development of the degradable RB-based polymeric products.

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References

- Borghei M., Karbassi A., khoramnejadian S., Oromiehie A., Javid A.H., 2010, Microbial biodegradable potato starch based low density polyethylene, African Journal of Biotechnology 9 (26), 4075-4080.
- Bourtoom T., Chinnan M.S., 2008, Preparation and properties of rice starch–chitosan blend biodegradable film, LWT Food Science and Technology 41 (9), 1633-1641.
- Garg S., Jana A.K., 2006, Effect of Starch Blends on Mechanical Properties of Low Density Polyethylene (LDPE) Film, Indian Chemical Engineering 48 (3), 185-189.
- Hu G., Huang S., Cao S., Ma Z., 2009, Effect of enrichment with hemicellulose from rice bran on chemical and functional properties of bread, Food Chemistry 115 (3), 839-842.
- Khoramnejadian S., 2011, Study of water absorption and biodegradation of potato starch based polyethylene, Advances in Environmental Biology 5 (10), 3156-3158.
- Moghadam R.A., Yusup S., Lam H. L, Shoaibi A.A., Ahmad M.M., 2013, Hydrogen Production from Mixture of Biomass and Polyethylene Waste in Fluidized Bed Catalytic Steam Co-Gasification Process, Chemical Engineering Transaction 35, 565-570.
- Monica P., Seggiani M., Vitolo S., 2015, Polyethylene and Hydrolyzed Collagen Blend Films Produced by Blown Extrusion, Chemical Engineering Transactions 43, 1705-1710.
- Morreale M., Scaffaro R., Maio A., La Mantia F.P., 2008, Effect of adding wood flour to the physical properties of a biodegradable polymer, Composites Part A: Applied Science and Manufacturing 39 (3), 503-513.
- Rahman W.A., Isa N.M., Rahmat A.R., Adenan N., Ali R.R., 2010, Rice husk/high density polyethylene biocomposite: effect of rice husk filler size and composition on injection molding processability with respect to impact property, Advanced Materials Research 83-86, 367-374.
- Sabetzadeh M., Bagheri R., Masoomi M., 2012, Effect of Corn Starch Content In Thermoplastic Starch/Low-Density Polyethylene Blends on Their Mechanical and Flow Properties, Journal of Applied Polymer Science 126, 63-69.
- Seggiani M., Cinelli P., Verstichel S., Puccini M., Vitolo S., Anguillesi I., Lazzeri A., 2015, Development of Fibres-Reinforced Biodegradable Composites, Chemical Engineering Transactions 43, 1813-1818.
- Soni D., Saiyad M., 2011, Biodegradable polyethylene bags, Proceeding of International Conference on Current Trends in Technology, 8-10 December 2011, Institute of Technology, Nirma University, 382-481.
- Tudorachi N., Cascaval C.N., Rusu M., Pruteanu M., 2000, Testing of polyvinyl alcohol and starch mixtures as biodegradable polymeric materials, Polymer Testing 19 (7), 785-799.
- Wang S., Yu J., Yu J., 2005, Compatible Thermoplastic Starch/Polyethylene Blends By One-Step Reactive Extrusion, Polymer International 54 (2), 279-285.